

REMARKS

Claims 1, 3, 4, 7 and 18 have been amended. Upon entry of this amendment, claims 1-29 will be pending in the application.

The specification has been amended at page 19 in response to the Examiner's objection set out at page 9 of the Office action.

* Attached hereto is a marked-up version of the changes made to the claims and specification by this amendment. The attached pages are captioned "Version With Markings to Show Changes Made."

Information Disclosure Statements

Applicants request that a copy of the initialed Form PTO/SB/08A accompanying the First and Second Supplemental Information Disclosure Statements filed October 9 and 21, 2002, respectively, be returned with next communication from the Patent Office in connection with this application.

Response to Restriction Requirement

The Examiner's courteous telephone interview with the undersigned attorney on January 14, 2003 is appreciated.

As discussed with the Examiner, claim 1 has been amended to more clearly recite that by-products containing reactive carbonyl groups (e.g., aldehydes and ketones such as n-butyraldehyde and butan-2-one) in the portion of the liquefiable products of the intermediate reaction product mixture introduced into the selective hydrogenation zone are selectively hydrogenated to corresponding alcohols (e.g., n-butanol and 2-butanol, respectively). See, for example, applicants' specification beginning at page 13, line 12. By contrast, the Grootendorst, et al. reference cited by the Examiner describes reduction of a carboxyl-containing compound, acetic acid (not a compound containing a reactive carbonyl group), to acetaldehyde (not an alcohol). Accordingly, step (c) in claim 1 as amended, constitutes a special technical feature common to all four inventions (Groups I-IV) previously identified by the Examiner.

During the aforementioned telephone interview, it was agreed that in light of this clarifying amendment, the restriction

requirement is no longer proper. Accordingly, applicants request that the restriction requirement be withdrawn.

Rejections under 35 U.S.C. §112, Second Paragraph

Applicants respectfully request reconsideration of the rejections of claims 1-29 under 35 U.S.C. §112, second paragraph.

It is clear that the "selective hydrogenation conditions" recited in step (c) of claim 1 as now amended must be "effective to selectively hydrogenate said by-products containing reactive carbonyl groups to corresponding alcohols." That is, the conditions selected are such that by-products containing reactive carbonyl groups are reduced to the corresponding alcohols preferentially over other reducible components of the liquefiable products of the intermediate reaction mixture (e.g., ethyl acetate).

Applicants further respectfully submit that omission of particular conditions encompassed by the term "selective hydrogenation conditions" does not render this term indefinite. To the contrary, conditions effective to selectively and preferentially hydrogenate by-products containing reactive carbonyl groups (e.g., aldehydes and ketones such as n-butylaldehyde and butan-2-one) in the portion of the liquefiable products of the intermediate reaction product mixture introduced into the selective hydrogenation zone to corresponding alcohols (e.g., n-butanol and 2-butanol, respectively) are readily ascertained by those of ordinary skill in the art, particularly in view of the exemplary selective hydrogenation conditions provided by applicants in their specification (See, for example, page 14, line 3 to page 15, line 4; and page 22, line 20 to page 23, line 23 of applicants' specification). Further delineation of the term "selective hydrogenation conditions" in claim 1 is unnecessary to distinguish the prior art and would unduly limit the protection to which applicants are entitled.

Applicants respectfully submit that the Examiner's rejection of claims 3, 4, and 7 as being indefinite for specifying that

certain ratios are "used" has been obviated by the present amendments to these claims.

Applicants further submit that the quotation marks in connection with the term "light" in original claims 18 was intended to indicate that this term is defined in applicants' specification. Claim 18 has been amended by removing the quotation marks and including the definition of "light" components found on page 17, lines 29-31 of applicants' specification.

In view of the foregoing, applicants respectfully request withdrawal of all claim rejections under 35 U.S.C. §112.

Rejections under 35 U.S.C. § 103(a)

Reconsideration is respectfully requested of the rejection of claims 1-29 under 35 U.S.C. §103(a). The invention defined in the pending claims is submitted as patentable over the disclosure of U.S. Patent No. 2,027,182 (Lazier).

The process of the present invention is directed to the production of ethyl acetate by the conversion (e.g., dehydrogenation) of a C₂ feedstock comprising ethanol. The recovery of substantially pure ethyl acetate is difficult due to the formation of byproducts having boiling points close to that of ethyl acetate (e.g., aldehyde and ketone byproducts containing reactive carbonyl groups such as n-butyraldehyde and butan-2-one). However, applicants have solved this problem by taking an intermediate reaction product mixture comprising hydrogen and liquefiable products comprising ethyl acetate, ethanol and by-products containing reactive carbonyl groups and contacting at least a portion of the liquefiable products of this reaction mixture with a selective hydrogenation catalyst in the presence of hydrogen in a selective hydrogenation zone to selectively hydrogenate the by-products containing reactive carbonyl groups to corresponding alcohols. That is, the by-products containing reactive carbonyl groups are reduced to the corresponding alcohols preferentially over other reducible components of the

liquefiable products of the intermediate reaction mixture (e.g., ethyl acetate).

The Examiner's indication of allowable subject matter on page 8 of the Office action is acknowledged. Applicants respectfully submit that step (c) of claim 1 as amended now clearly specifies that by-products containing reactive carbonyl groups (e.g., aldehydes and ketones such as n-butyraldehyde and butan-2-one) in the portion of the liquefiable products of the intermediate reaction product mixture introduced into the selective hydrogenation zone are selectively hydrogenated to corresponding alcohols (e.g., n-butanol and 2-butanol, respectively). Accordingly, as indicated in the Office action, the claim rejections under 35 U.S.C. §103(a) are obviated by the instant amendment.

Applicants acknowledge that Lazier discloses the production of ethyl acetate by the catalytic dehydrogenation of ethanol (i.e., step (a)(i) of the claimed invention). More particularly, the object of Lazier is to produce higher alcohols and their esters by hydrogenating olefinically unsaturated alcohols and esters obtained in the catalytic dehydrogenation of ethanol to form ethyl acetate. However, in contrast to the claimed invention, this reference fails to teach or suggest selective hydrogenation of the effluent of the dehydrogenation of ethanol and, accordingly, fails to teach or suggest the process defined in claim 1 as amended, including step (c) in which at least a portion of the liquefiable products of the intermediate reaction product mixture is contacted with a selective hydrogenation catalyst in a selective hydrogenation zone in which by-products containing reactive carbonyl-containing groups are hydrogenated to produce the corresponding alcohols.

The effluent resulting from the dehydrogenation of ethanol as taught by Lazier is said to contain ethyl acetate, acetaldehyde, acetone, butanol, crotonyl alcohol, butyl acetate, crotonyl acetate, acetic acid, butyric acid, crotonic acid, caprylic acid and capric acid. The acetic acid, butyric acid, crotonic acid, caprylic acid and capric acid, as well as the

esters butyl acetate and crotonyl acetate, present in the effluent produced by Lazier's dehydrogenation of ethanol each contain a carboxyl group, not a reactive carbonyl group as found in aldehydes and ketones. Of the remaining components of the effluent, butanol and crotonyl alcohol clearly do not contain a carbonyl group, while only acetone does. Importantly, based on the boiling points of the components of the dehydrogenation effluent of Lazier, there is no compound and, in particular, no reactive carbonyl containing compound, present which is likely to form an azeotrope with ethyl acetate.¹ This is in stark contrast to applicants' disclosed invention directed to the situation in which the by-products formed in the dehydrogenation of a C₂ feedstock comprising ethanol to ethyl acetate have boiling points close to that of ethyl acetate such that recovery of substantially pure ethyl acetate from the esterification product mixture is complicated. Thus, Lazier fails to even contemplate the particular problems addressed by the present invention, much less teach or suggest the solution defined in the pending claims.

The hydrogenation step as taught by Lazier is not conducted on the dehydrogenation effluent, but rather on crude synthetic higher alcohols separated from the effluent by distillation. Moreover, there is absolutely no mention in Lazier of selectively hydrogenating reactive carbonyl-containing by-products of the dehydrogenation of ethanol and, accordingly, no teaching or

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Ethyl acetate	77°
Acetaldehyde	20.2°
Acetone	56.2°
1-butanol	117.7°
Crotyl alcohol	121-126°
Butyl acetate	126.3°
Crotyl acetate	120 - 125°C*
Acetic acid	118°
Butyric acid	163.5°
Crotonic acid	185°
Caprylic acid	237.9°

Unless otherwise indicated, boiling point data was obtained from Hawley's Condensed Chemical Dictionary, 14th Ed. (* U.S. Patent No. 6,456,683)

suggestion of selective hydrogenation of reactive carbonyl-containing components to the corresponding alcohols.

Applicants point out that Lazier contains no restriction of the hydrogenation catalyst to those that will not significantly hydrogenate esters whereas the present application describes a "selective hydrogenation catalyst" as having "good activity for hydrogenation of reactive carbonyl containing compounds, but relatively poor ester hydrogenation activity" (See, page 14, line 16 et seq. of applicants' specification). Applicants also direct the Examiner's particular attention to Example 5 of Lazier in which a copper chromite catalyst is used at 225°C and 3000 psi (207 bar) hydrogen pressure for hydrogenation of crude synthetic higher alcohols obtained by high pressure dehydrogenation of ethanol. At these conditions, any ethyl acetate present would be substantially hydrogenated to ethanol. This is completely contrary to the selective hydrogenation step recited in step (c) of claim 1 in which byproducts containing reactive carbonyl groups are selectively hydrogenated to the corresponding alcohols over other reducible components of the liquefiable products such that ethyl acetate are unaffected by the selective hydrogenation conditions.

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In view of the above, applicants respectfully submit that Lazier fails to teach disclose or suggest the process defined in claim 1, including the selective hydrogenation step recited in step (c), such that a *prima facie* case of obviousness is lacking. Accordingly, claim 1 and dependent claim 2-29 are submitted as patentable over Lazier.

Conclusion

Favorable reconsideration and allowance of all pending claims are respectfully solicited.

Applicants request an extension of time to and including February 10, 2003 for filing a response to the above-mentioned Office action. A check in payment of the applicable extension fee is enclosed.

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The Commissioner is requested to charge any fee deficiency of overpayment in connection with this amendment to Deposit Account 19-1345.

Respectfully submitted,



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VERSION WITH MARKINGS TO SHOW CHANGES MADE

IN THE SPECIFICATION:

The following heading has been inserted between the paragraph beginning at page 19, line 10 and the paragraph beginning at page 19, line 13:

Brief Description of the Drawings

IN THE CLAIMS:

Claim 1 has been amended as follows:

1. (once amended) A process for the production of ethyl acetate which comprises:

(a) converting a C₂ feedstock comprising ethanol to ethyl acetate in an ethyl acetate production zone by a procedure selected from:

- (i) dehydrogenation,
- (ii) oxidation,
- (iii) reaction with acetaldehyde, and
- (iv) oxidation to acetaldehyde followed by the Tischenko reaction;

(b) recovering from the ethyl acetate production zone an intermediate reaction product mixture comprising hydrogen and liquefiable products comprising ethyl acetate, ethanol, and by-products containing reactive carbonyl groups;

(c) contacting at least a portion of the liquefiable products of the intermediate reaction product mixture with a selective hydrogenation catalyst in the presence of hydrogen in a selective hydrogenation zone maintained under selective hydrogenation conditions effective [for selective hydrogenation of by-products containing reactive carbonyl groups thereby] to selectively hydrogenate said by-products [selectively] containing reactive carbonyl groups to [hydrogenated by-products comprising] corresponding alcohols;

(d) recovering from the selective hydrogenation zone a selectively hydrogenated reaction product mixture comprising ethyl acetate, ethanol, hydrogen and hydrogenated by-products comprising said corresponding alcohols;

(e) distilling material of the selectively hydrogenated reaction product mixture in one or more distillation zones so as to produce a first composition comprising substantially pure ethyl acetate and a second composition comprising ethanol and water;

(f) treating the second composition of step (e) to separate water therefrom and yield a third composition comprising ethanol with a reduced water content; and

(g) recovering the third composition of step (f).

Claim 3 has been amended as follows:

3. (once amended) A process according to claim 2, wherein the **[dehydrogenation conditions include use of an]** ethanol:hydrogen molar ratio **[of] in the dehydrogenation zone is** from about 1:10 to about 1000:1, **[a] the** combined partial pressure of ethanol and hydrogen **[of] in the dehydrogenation zone is** from about 3 bar (3×10^5 Pa) up to about 50 bar (5×10^6 Pa), and **[a] the** temperature **in the dehydrogenation zone is** **[in the range of]** from about 100°C to about 260°C.

Claim 4 has been amended as follows:

4. (once amended) A process according to claim 3, wherein the **[dehydrogenation conditions include use of a]** combined partial pressure of ethanol and hydrogen **[of] in the dehydrogenation zone is** at least about 6 bar (6×10^5 Pa) up to about 30 bar (3×10^6 Pa).

Claim 7 has been amended as follows:

7. (twice amended) A process according to claim 1, wherein the selective hydrogenation conditions **in the selective hydrogenation zone** of step (c) include **[use of]** a reaction product mixture:hydrogen molar ratio of from about 1000:1 to about 1:1, a combined partial pressure of the liquefiable

products of the intermediate reaction product mixture and hydrogen of from about 5 bar (5×10^5 Pa) to about 80 bar (8×10^6 Pa), and a temperature in the range of from about 20°C to about 160°C.

Claim 18 has been amended as follows:

18. (twice amended) A process according to claim 12, wherein an ethanol rich stream containing substantially all of the water in the selectively hydrogenated reaction product mixture is recovered from the bottom of the first distillation zone, while an overhead stream that contains ["light"] light components having lower boiling points than ethyl acetate and its azeotropes with water and ethanol present in the selectively hydrogenated reaction product mixture is recovered from the first distillation zone, and in which the first distillate comprises a liquid draw stream which is recovered from an upper region of the first distillation zone and which comprises ethyl acetate, ethanol, water and minor amounts of other components.